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⑤⑥ References cited:
EP-A- 0 165 111
US-A- 4 033 673
US-A- 4 550 982

DISPLAY & IMAGING TECHNOLOGY, vol. 1,
no. 1, 1985, pages 61-80, Gordon and Breach
Science Publishers Ltd and OPA Ltd; Y.
HIRAI et al.: "Dynamic characteristics analy-
sis for redox-pair electrochromic display
based on diffusion-annihilation model"

PATENT ABSTRACTS OF JAPAN, vol. 11, no.
342 (P-365)[2789], 10th November 1987, page
17 P 635;& JP-A-62 124 534

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Description

This invention relates to an organic thin film display element.

In recent years, the development of the technique of forming an extremely thin film of organic material represented by Langmuir-Blodgett process (hereinafter referred to as "LB process") has lead to the active application of said technique to various electronic elements.

Studies in this particular field (represented, for example, by the research work on the MIS type semiconductor element using a thin organic film) are widely pursued. Under the present state, however, no new functional element has been practically proposed which effectively utilizes the properties peculiar to said thin organic film.

Among the properties of the organic material, that which draws the greatest attention from the view point of applying said material to a functional element is a charge transfer phenomenon among the molecules. In this connection it will be noted that some of the organic materials comprise donor molecules which have a low ionization potential and tend to be converted into positive ions by supplying electrons to other molecules. The other organic materials are formed of acceptor molecules which have a high electron affinity and receive electrons from other molecules and tend to be negative ions by themselves. It is well known that these two types of molecules are combined into a compound generally referred to as a charge-transfer complex. For instance a complex of perylene and tetracyanoquinodimethane (TCNQ) is formed of neutral molecules giving rise no charge transfer. In contrast, a complex of tetramethylphenylenediamine (TMPD) and TCNQ represents an ionic compound composed of positive and negative molecules. It is also known that a compound of tetrathiofulvalene (TTE) and TCNQ indicates a transition from a neutral to an ionic state depending on temperature and pressure.

When the charge transfer phenomenon of the above-mentioned organic material is utilized as an operation principle of an element, it is required that the organic material be possessed of excellent charge transfer properties including quick responsibility and high controllability, and further can be easily prepared. However, the satisfaction of the above-mentioned requirements is accompanied with the drawbacks that the charge-transfer complex crystal presents tremendous difficulties in the growth of a crystal, and it is difficult to externally control the charge transfer. Further, the control of a charge transfer between a metal thin film and organic thin film by means of a light or electric field is applied to a switching element or memory element. However, this attempt is accompanied with great drawbacks in respect of the charge transfer efficiency, the speed of responsibility, the effective life of the processed elements, etc. As mentioned above, the phenomenon of charge transfer in an organic material has not yet been fully put to practical application, though great hope has been pinned on the utilization of the charge transfer phenomenon to the organic element.

Another noticeable property of an organic material in question is its optical characteristic. Like various organic coloring materials, many of the proposed organic materials respond to light falling within the visible range and some are possessed of a high light-absorbing property. To date, an electro-chromic display (ECD) element has been known as a element to which the optical property of the aforementioned organic material has been applied. This ECD element is characterized in that the ionic condition of an organic material is changed by impressing an electric field on organic molecules in a solution, and the display of an image is carried out by changes in the absorption spectrum occurring in the visible range. The ECD element offers the advantages that an image is distinctly displayed and the view field is very much broadened.

Nevertheless, the ECD element is still accompanied with the drawbacks that the behavior of the ECD is followed by the shifting of ions or protons, resulting in an extreme decline in the responding speed and operating efficiency. Moreover, the ECD element presents difficulties in the repeating characteristic and moreover has a short effective life. Consequently, the ECD element is retarded in practical application in spite of its excellent optical properties.

Further, a great demand is cried for the development of a new element which is possessed of such a novel function as can not be expected from the conventional electroluminescence device, light-emitting diode (LED), liquid crystal display element, etc. Such a novel function may be represented, for example, by that of a single picture element capable of displaying various tints. A light-emitting display element represented by the conventional LED has the drawback that its light-emitting property is defined by the parameter of the raw material of said element. Tremendous difficulties are encountered in controlling the wavelength of an emitted light by an external factor such as voltage. Moreover, a liquid crystal display element indicates colors by means of filters of red, green and blue colors. Essentially, therefore, one picture element of such an element is capable of displaying only one color.

US-A-4033673 discloses an organic thin film display element with an organic thin film containing donor

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molecules and acceptor molecules. The thin film is positioned between a pair of electrodes parallel to the film. Between the thin film and each electrode there is provided a membrane.

It is accordingly an object of the present invention to provide a display element using an organic thin film having excellent properties in respect of responding speed, efficiency and effective life.

5 Another object of the invention is to provide a thin organic film color display device in which a single picture element is capable of displaying a plurality of colors one after another.

According to this invention, there is provided an organic thin film display element comprising: an organic thin film containing at least two parallel organic monomolecular layers, one of which contains organic molecules of at least the donor kind, the other of which contains organic molecules of at least the
10 acceptor kind; a pair of electrodes parallel to said thin film, said thin film being positioned between said electrodes; at least a first additional monomolecular layer containing insulating organic molecules and being positioned between one of said electrodes and said thin film; at least a second additional monomolecular layer containing insulating organic molecules and being positioned between the other of said electrodes and said thin film; said electrodes being arranged to cause a charge transfer between the donor molecules and
15 the acceptor molecules, said charge transfer varying the optical transmission of the organic thin film, whereby the organic thin film displays an image.

Charge transfer between said donor molecules and acceptor molecules is effected by impressing a voltage between the electrodes on a thin organic film. The thin organic film may be composed by alternately and repeatedly laminating a layer containing donor molecules and a layer containing acceptor
20 molecules. In these cases, a layer containing insulating organic molecules may be interposed between the said layers. The layer containing insulating organic molecules may be interposed between pairs of donor molecule and acceptor molecule layers. Said insulating organic molecules may also be mixed with the donor molecules, as well as with the acceptor molecules.

As used herein, the donor molecule is defined to mean the organic molecule which has a small
25 ionization potential (I_p) and releases electrons to another molecule and is readily turned into a positive ion. The above-mentioned donor molecules related to the present invention may be listed as below:

(1) Fulvalene type donors having the undermentioned structural formulas:

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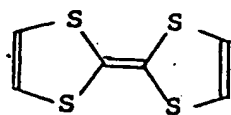
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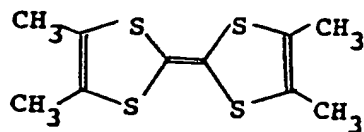
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TTF

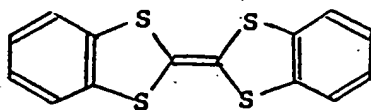
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TMTTF

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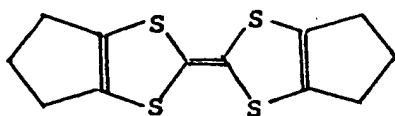
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DBTTF

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HMTTF

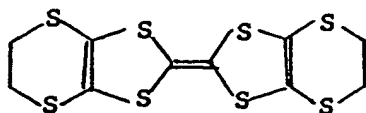
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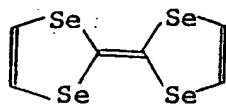
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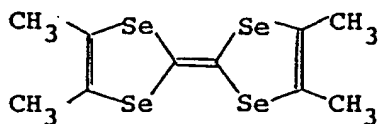


BEDT-TTF

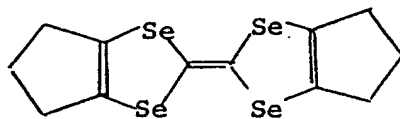
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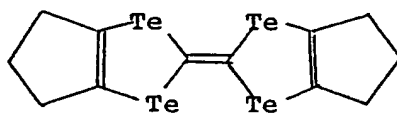
TSF



TMTSF

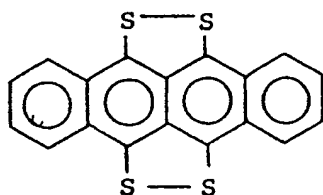
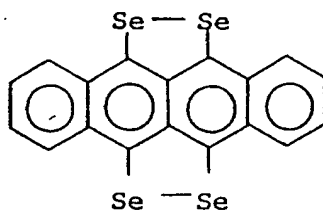


HMTSF



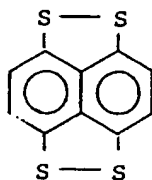
HMTTeF

(2) S-containing heterocyclic donor having the undermentioned structural formulas:

tetrathiotetracene
(TTT)tetraselenatetracene
(TST)

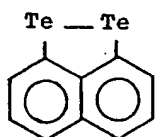
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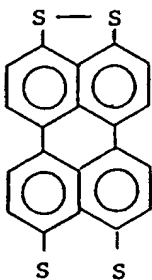
tetrathionaphthalene

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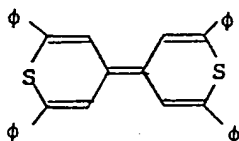
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tetrathioperillene

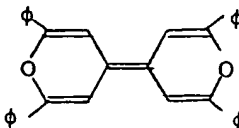
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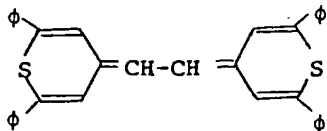
tetraphenyl-bithio pyralydene
(BTP)

40



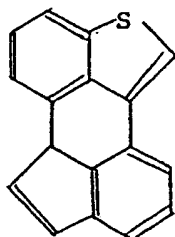
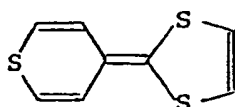
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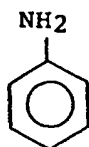


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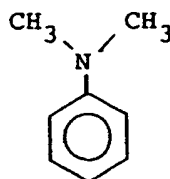
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bizenzothiophene
(BBT)Note: ϕ given in the above formulas represents a phenyl radical.

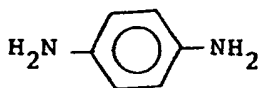
(3) Amine type donor having the undermentioned structural formulas:



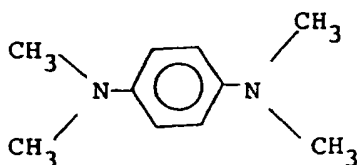
aniline



N-methyl aniline

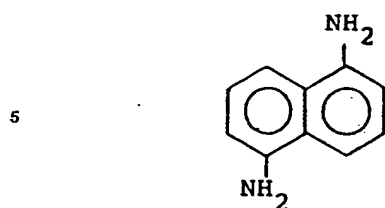


p-phenylenediamine

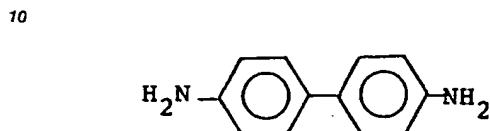


TMPD

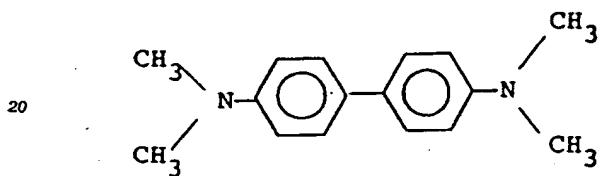
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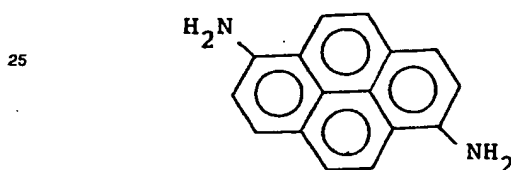
diaminonaphthalene



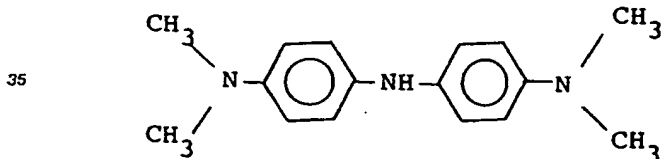
benzidine



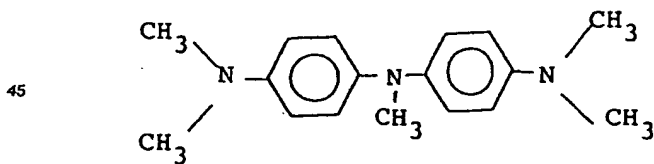
tetramethyl benzidine



diaminopyrene



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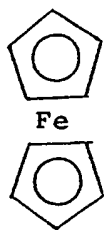


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(4) Metal compound type donor having the undermentioned structural formulas:

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ferrocene

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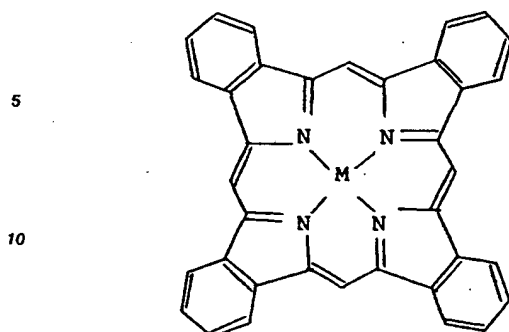
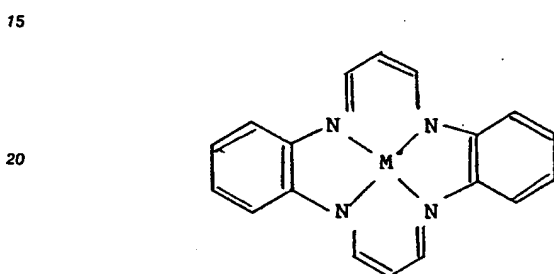
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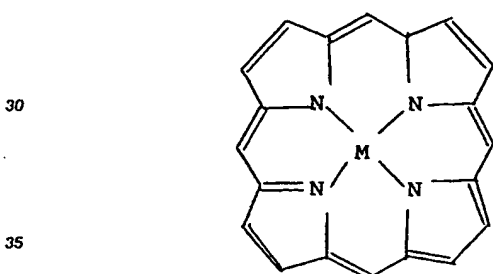
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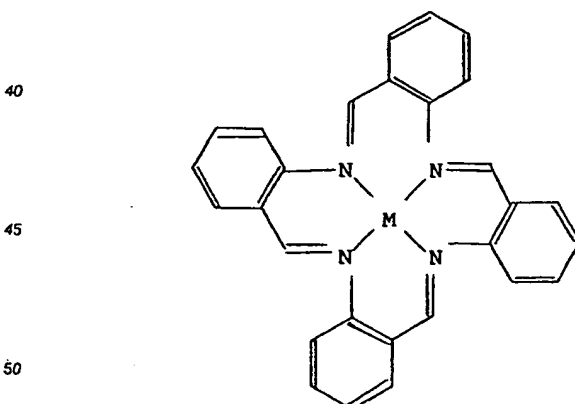
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phthalocyanine
M(Pc)

Ni(dbtaa)



porphyrin

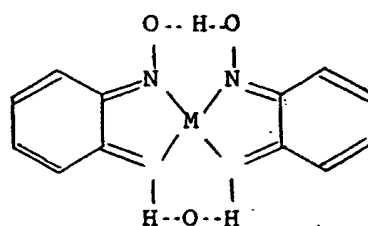


M(TAAB)

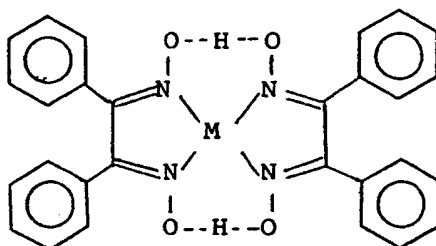
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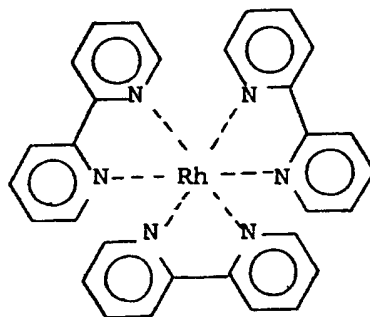
 $M(bqd)_2$

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 $M(dpq)_2$

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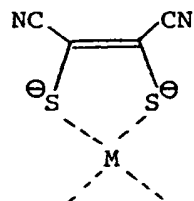
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 $Rh(bpy)_3$

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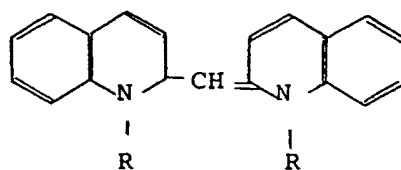


MNT

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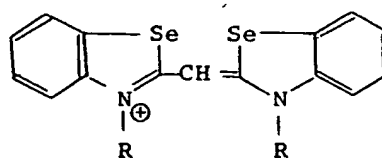
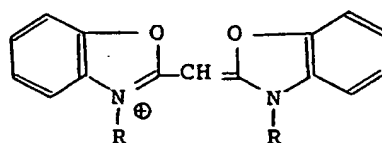
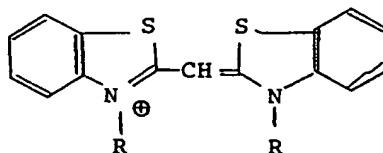
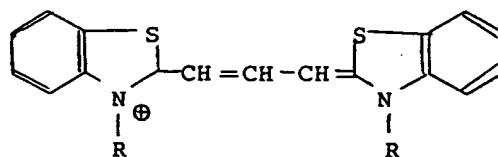
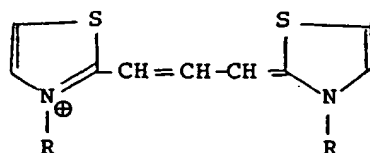
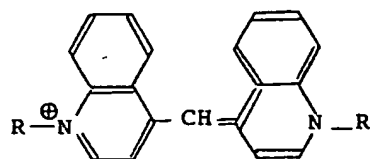
(5) Cyanin coloring matter donor having the undermentioned structural formulas:

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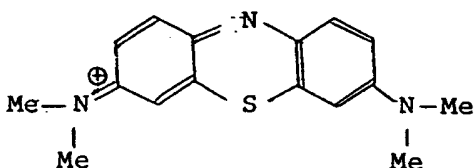
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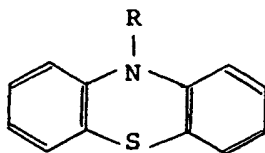
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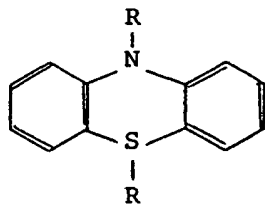
(6) N-containing heterocyclic donor having the undermentioned structural formulas:

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phenothiazine

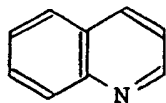
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phenazine

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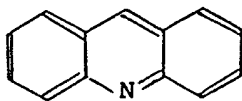
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quinoline

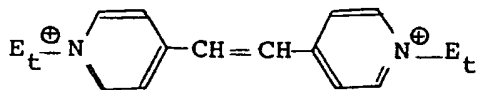
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acridine

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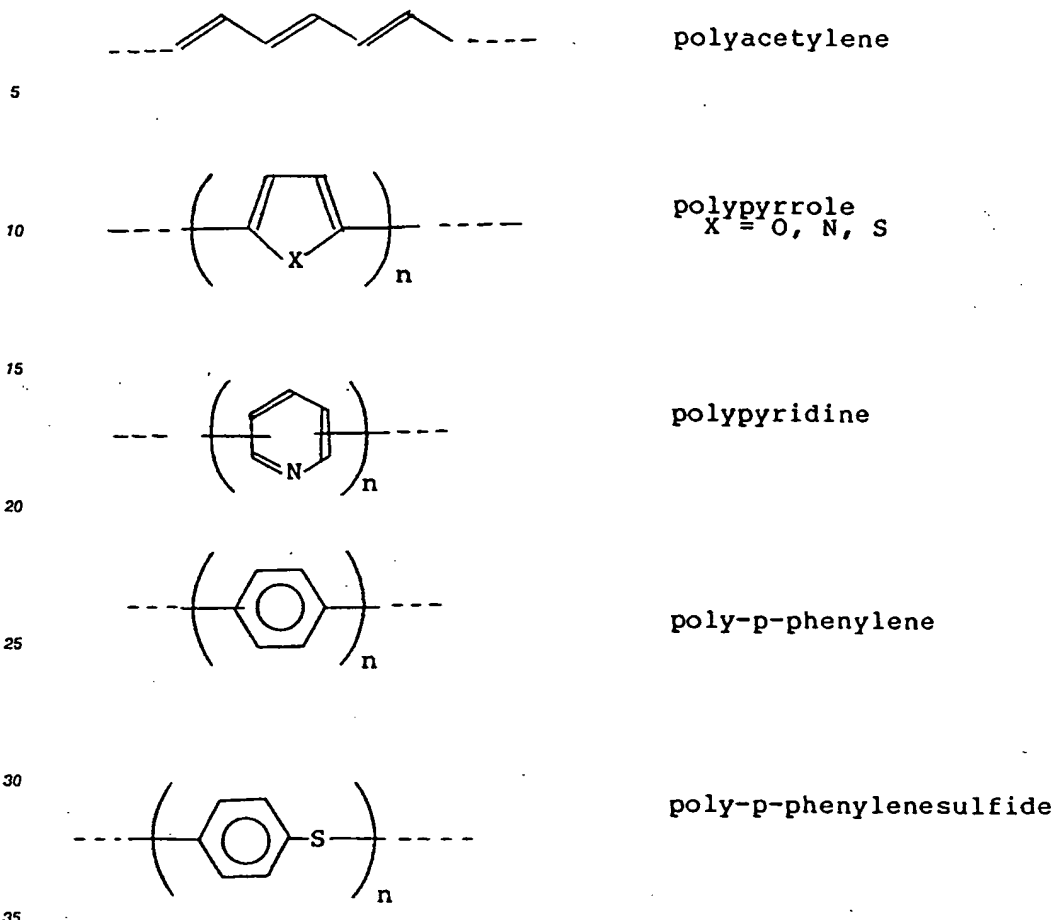
DEPE²⁺

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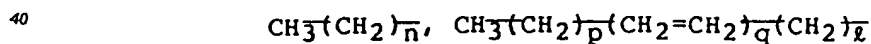
(7) Polymer type donor having the undermentioned structural formulas:

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The present invention allows for the application of the derivatives of the donor type molecules indicated in items (1) to (7), which have hydrophobic group with a structural formula



(where n and p+q+l respectively represent larger numbers than 8), or hydrophilic group with a structural formula of -COOH, -OH, SO₃H, -COOR', -NH₂, -N^o(R')₃Y⁻ (Y denotes halogens), or have both said

45 hydrophobic and hydrophilic groups.

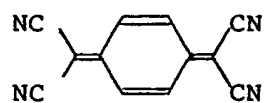
Among the aforementioned donor molecules, amine type donors are preferred, but fulvalene type donors and S-containing heterocyclic compound type donors are more preferred.

As used herein, the term "acceptor molecule" is defined to mean an organic molecule which has a great electron affinity (E_A), and tends to receive electrons from another molecule and be readily converted

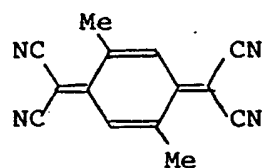
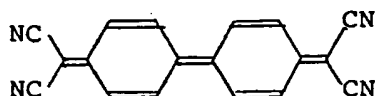
50 into negative ions. The above-defined acceptor molecules include the following:

(1) Cyano compound type acceptors having the undermentioned structural formulas:

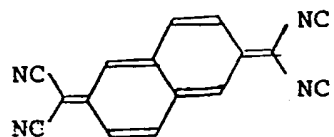
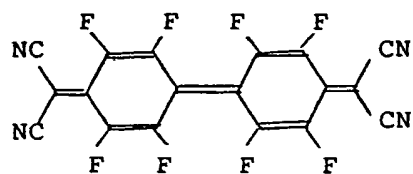
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TCNQ

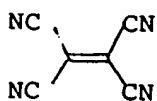
Me₂TCNQ

TCNDQ

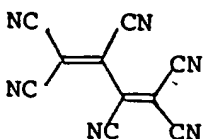


TNAP

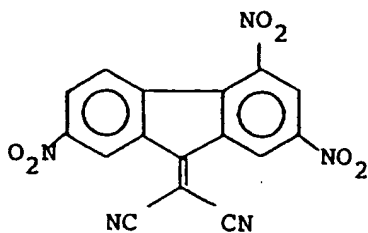
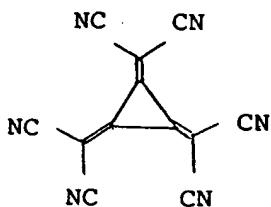
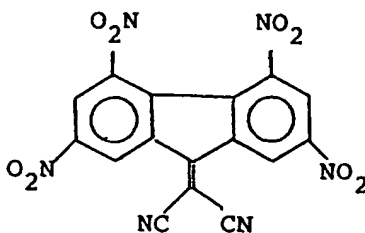
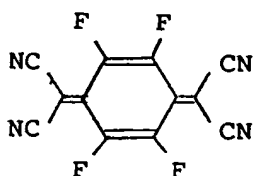
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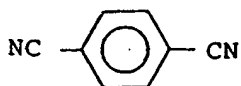
TCNE



HCB



DTF

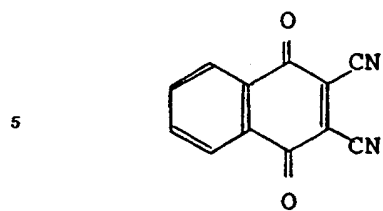


dicyanobenzene

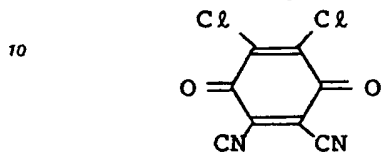
DCNB

(2) Quinone type acceptor having the undermentioned structural formulas:

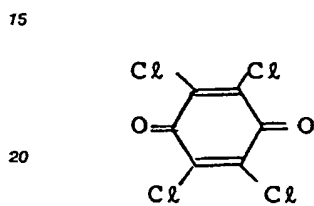
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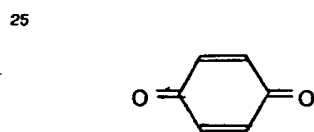
dicyanonaphthoquinone



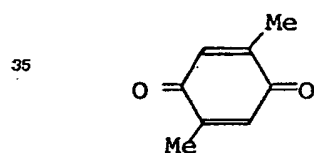
DDQ



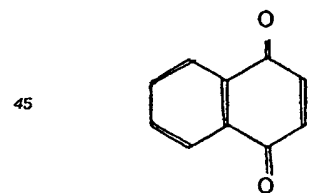
chloranil



benzoquinone



2,5-dimethylbenzoquinone

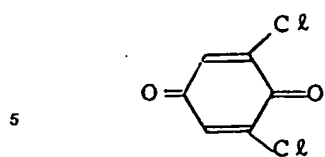


naphthoquinone

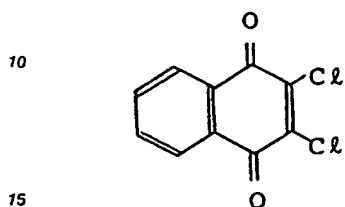
50

55

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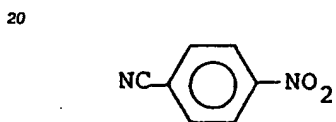


2,6-dichlorobenzoquinone

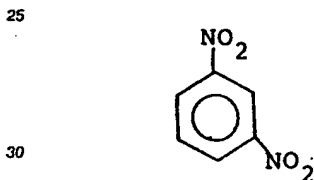


2,3-dichloronaphthoquinone

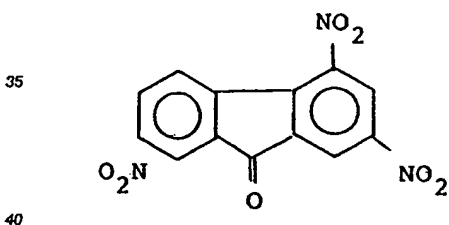
(3) Nitro-compound type acceptor having the undermentioned formulas:



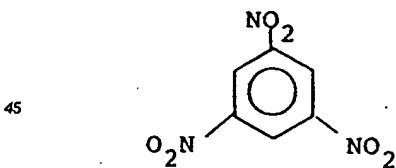
p-nitrocyanobenzene



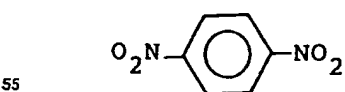
1,3-dinitrobenzene



TNF



1,3,5-trinitrobenzene

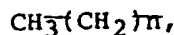


p-nitrobenzen

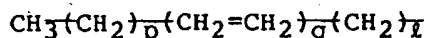
The present invention allows for the application of the derivatives of the acceptor type molecules having

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the structural formulas indicated in items (1) to (3), which have hydrophobic radicals represented by



5



10 (where n and a total of p+q+l respectively denote a larger number than 8), or hydrophilic radicals represented by -COOH, -OH, -SO₃H, -COOR', -NH₂, -N⁺(R')₃Y⁻¹ (Y denotes a halogen), or have both such hydrophobic and hydrophilic radicals.

Among the above-listed acceptor molecules quinone type donors are preferred and cyano compound type donors are more preferred. As used herein, the term "insulating organic molecule" is defined to mean 15 that molecule which does not exchange electrons with respect to other molecules, such as those which have a lower electron affinity and a higher ionization potential than the donor and acceptor molecules. The above-defined insulating organic molecules include the undermentioned types.

(1) Substitutable saturated or unsaturated hydrocarbon derivatives which are expressed by the following general structural formula:

20



where R represents a hydrophobic radical formed of substitutable

25



(where n and a total of p+q+l respectively represent a larger number than 8), and X represents hydrophilic radicals, including, for example, -COOH, -OH, -SO₃H, -COOR', -NH₂, -N⁺(R')₃Y⁻ (Y represents a halogen). 30

(2) Various polymerizable molecules

The subject polymerizable molecules may be represented, for example, by:

polymers of vinyl compound such as substitutable acrylate, methacrylate, vinyl ether, styrene, vinyl alcohol, acrylamide, acryl;

35 α-amino acids such as alanine, glutamate, aspartate;

amino acids, for example, ε-aminocaproate other than α-amino acid; and

polyamide polymers composed of a 1:1 mixture of diamines such as hexamethylene diamine and carbonic acid such as hexamethylene diamine.

If possessed of a property of being fabricated into a film by themselves, the above-mentioned 40 molecules (2) can be applied in tact. The molecules which can not be fabricated into a film by themselves are applied in the form mixed with the insulating molecules (listed in item (1)) which can be independently formed into a film.

It is preferred that the aforementioned first, second and third films be respectively applied in the form of a monomolecular film fabricated by the Langmuir Blodgett process or in the stacked form.

45 When a thin organic film display device embodying the present invention is provided with an electrode at both ends, and a bias voltage is impressed on said electrodes, the transfer of electrons or holes takes place between the electronic state of a donor molecule and that of an acceptor molecule. Namely, when the bias voltage exceeds a level expressed as

$$50 \{I_{pD} - E_A\} / e \quad [V]$$

where:

I_{pD} = ionization potential of a donor molecule

E_A = electron affinity of an acceptor molecule

55 $-e^2/a$ = coulomb potential appearing between the holes produced in the donor molecule and those generated in the acceptor molecule

then the electronic state of the donor molecule transits to that of the acceptor molecule. As a result of said transition, the donor molecule and acceptor molecule change in valence. At this time a noticeable change

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appears in the absorption spectrum of the visible section. The display element representing the present invention causes said change in the absorption spectrum to be utilized in a display function.

When the organic film is a laminated body comprising a first organic film containing donor molecules and a second organic film containing acceptor molecules, the transfer of electric charges takes place in an interstice between said first and second organic films. When an organic film formed of a mixture of donor molecules and acceptor molecules is a monomolecular type, the transfer of electric charges occurs between the donor and acceptor molecules involved in a monomolecular film. When the organic film consists of stacked monomolecular film, the transfer of an electric charge takes place between the donor and acceptor molecules contained in a monomolecular film as well as between the donor and acceptor molecules contained in the adjacent monomolecular films.

As described above, a display element embodying the present invention is characterized by the switching operation caused by the transfer of electrons or holes between the organic molecules. To minimize, therefore, variations in the switching voltage, it is necessary to control the thickness of the respective films to the order of Å. Further, the respective films are demanded to have a high uniformity of properties and be as much free from defects as possible. To attain these objects, therefore, it is preferred that the subject organic film be fabricated by the Langmuir-Blodgett process.

The first film containing donor molecules can be prepared, for example, by spreading a liquid containing a mixture of insulating organic molecules and donor molecules on the water for compression and then transferring the compressed mass to the substrate surface or by bonding a hydrophilic or hydrophobic radical with the donor molecules, spreading a solution consisting of said bonded molecules on the water for compression and transferring the compressed mass to the substrate.

The second film containing acceptor molecules can be fabricated by the same process as mentioned above.

The organic thin film display device of the present invention offers the following advantages.

First, since donor and acceptor molecules can be freely selected, it is possible to control the bias voltage for effecting the transfer of electrons or holes namely, switching voltage to a proper level. The light-absorbing property of the subject device accompanied with valency changes varies with the type of the respective molecules. Therefore, it is possible to display any visible blue, green or red color at will.

Secondly, the device of this invention has a different display arrangement from the conventional ECD. Namely, the transfer of electrons or holes has the merit of extremely accelerating a response speed, namely, giving a response in as short a time as about 1 nsec. Since the display operation does not give rise to any change in the molecular structure of the film, any repeated application of the subject display device does not lead to the deterioration of the property. Therefore, said device is enabled to have a long effective life.

In the third place, the subject display device has a super lattice structure attained by repetitively laminating monomolecular organic films, thus realizing a highly efficient operation. Namely, a state transition between the donor molecule and acceptor molecule noticeably arises at the junction. Since the above-mentioned super lattice structure has a large number of such junctions, the total charge transfer amount at a certain bias voltage is tremendously larger than at the junction between the donor molecule layer and acceptor molecule layer constituting, for example, a thick film. Therefore, the organic film display device of this invention has an extremely high light-absorbing efficiency per unit bias voltage.

Description has been made of the case where a single color display is performed with respect to one picture element. However, the present invention is characterized in that since either donor molecule or acceptor molecule is formed of several types, it is possible to display a large number of colors in accordance with the number of combination of the donor and acceptor molecules.

As far as the object of this invention is concerned, it will be sufficient if the subject organic film display device comprises a first donor molecule-containing film laminated with a second acceptor molecule-containing film, and either donor or acceptor molecule is formed of several types. Further, it is possible to let the subject display device have a super lattice structure by repeatedly superposing a plurality of laminated bodies comprised of first donor molecule-containing organic films and second acceptor molecule-containing organic films. Or it is possible to provide a large series of laminated films respectively containing both donor and acceptor molecules, with either donor or acceptor molecule designed to comprise a plurality of types. Further, a laminated body consisting of a first donor molecule-containing film and a second acceptor molecule-containing film may be assembled with another laminated body comprised of a third organic film containing a different type of donor from that of the first film and a fourth organic film containing a different type of acceptor from that of the second film. The organic film can be rendered useful if at least one plane of said film is provided with an organic film containing insulating molecules. Thus, when an organic film in which either donor or acceptor molecule is comprised of a plurality of types like the

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aforementioned arrangement is used, donor and acceptor molecules jointly realizing the transfer of electrons can be assembled in a plurality of combinations. If molecules are assembled in a different way, a notice charge appears in the level of voltage at which electron transfer takes place. If, in case voltage level is progressively changed, each time electron transfer takes place between the respective assemblies of donor molecules and acceptor molecules, then changes will take place in the light absorbing property of the organic film. If this light absorbing property of the organic film is utilized as a display function, one picture element can display different colors one after another, according as the voltage impressed on said picture element varies in magnitude.

This invention can be more fully understood from the following detailed description when taken in conjunction with the accompanying drawings, in which:

- Fig. 1 illustrates the principle by which the present invention displays a single color;
- Fig. 2 shows the principle by which the present invention displays a plurality of colors;
- Fig. 3 is a sectional view of a display element according to a first embodiment of this invention, wherein donor molecule films and acceptor molecule films are alternately laminated with an insulating film interposed therebetween;
- Fig. 4 is a band pattern illustrating the phenomenon of charge transfer occurring in the display element of Fig. 3;
- Figs. 5 and 6 are sectional views showing the arrangements of organic films used as display elements according to Examples 2 and 3 of the present invention;
- Figs. 7A and 7B indicate the matrix electrodes of a display element according to Example 4 of the invention;
- Figs. 8A and 8B are timing charts indicating the operation of the display element according to Example 4 of the invention;
- Fig. 9 is a sectional view of a multicolor display element according to Example 5 of the invention;
- Fig. 10 illustrates the spectral transmittance curves shown by the display element according to Example 5 of the invention;
- Figs. 11 and 12 are sectional views of the multicolor display elements according to Examples 6 and 7 of the invention; and
- Figs. 13A and 13B are timing charts illustrating the operation of the multicolor display element according to Example 8 of the invention.

Description is first made of the phenomenon of charge transfer in the laminate structure consisting of a first organic film containing donor molecules and a second organic film containing acceptor molecules. The operation of the display element of the present invention is fundamentally based on the charge transfer occurring in the laminated body of said first and second films.

Fig. 1 shows an arrangement consisting of first and second films 13, 14 having hetero structures which are interposed between two electrodes 11, 15. A third film 12a containing insulating molecules is interposed between electrode 11 and second film 14. Another third film 12b containing insulating molecules is intervened between electrode 15 and first film 13. Said third films 12a, 12b are provided as barriers for preventing charge transfer between electrode 11 and second film 14 as well as between electrode 15 and first film 13.

According to the display element of the present invention arranged as described above, voltage is impressed across electrodes 11, 15 so as to cause the side of electrode 11 to be rendered positive. When said voltage becomes higher than voltage V_{tr} indicated by the undermentioned formula:

$$V_{tr} = \frac{l}{a} \cdot \frac{1}{e} \left\{ (I_{PD} - E_A - \frac{e^2}{a}) \right\} \quad \dots (1)$$

electrons transit from first film 13 to second film 14, thereby causing the donor molecules to have a positive ion (D^+) and the acceptor molecules to have a negative ion (A^-).

In the above formula, I_{PD} represents the ionized potential of the donor molecule, and E_A denotes the electron affinity of the acceptor molecule. l shows a distance between electrodes 11, 15. a indicates a distance between the hole of the donor molecule and the electron of the acceptor molecule. e denotes a charge elementary quantity.

Molecules absorb light beams to noticeably different extents according as they assume a neutral or ionized state. When electron transfer takes place, for example, from the donor molecule to the acceptor molecule, a tremendous change appears in the light absorption spectrum of the organic film having the structure illustrated in Fig. 1.

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Description may now be made of the phenomenon of charge transfer which occurs when a multicolor display is performed when either donor molecule or acceptor molecule is comprised of a plurality of kinds. Fig. 2 represents the case where first film 13 is composed of two different donor molecules D_1 , D_2 . Now let it be assumed that the ionized potential of the donor molecule is represented by $I_p(D_1) < I_p(D_2)$, and further
 5 the concentration of the acceptor molecular is equal to the total concentration of two donor molecules. Further, threshold voltages $V_{tr}(1)$ and $V_{tr}(2)$ shown in Formula (1) are assumed to correspond to two assemblies of donor and acceptor molecules $\{D_1, A\}$, $\{D_2, A\}$. If a voltage impressed across the two electrodes 11, 15, is higher than $V_{tr}(1)$, electron transfer arises between D_1 and A. If the impressed voltage rises higher than $V_{tr}(2)$, electron transfer appears between D_2 and A. The above-mentioned arrangement,
 10 therefore, gives rises to three absorption spectrum patterns, namely, one neutral state and two ionized state. When changes in the absorption spectra are utilized as a color display function, it is possible to cause a single picture element to display a plurality of colors.

The present invention will become more apparent by reference to the examples which follow.

15 Example 1

Fig. 3 is a sectional view of the display element according to the first embodiment of the invention. Reference numeral 21 denotes a glass substrate 21. A transparent conductive film 22 is formed on the surface of said substrate 21. 10 to 100 layers of first donor molecule-containing films 23 and second
 20 acceptor molecule-containing films 24 are alternately laminated with an insulating organic film 25 interposed between said first and second organic films 23, 24, thereby providing a super lattice structure. These films are fabricated by the Langmuir-Blodgett process. A translucent gold electrode 26 is thermally deposited in vacuum on the top of said super lattice structure with a thickness of 50 nm. Though not shown in Fig. 3, a gold lead having a diameter of 0.3 mm is fitted by means of silver paste of gold electrode 26 and
 25 transparent electrode 22 deposited on the substrate. The concrete process of the aforementioned super lattice structure involves the undermentioned steps. 100 μ l of a solutions is prepared by dissolving 500 mg of arachic acid ($CH_3(CH_2)_{18}COOH$) and 300 mg of tetratiafulvalene (TTF) in 100 ml of a solvent prepared by mixing chloroform and acetonitrile in the volumetric ratio of 1:1. Said solution of 100 μ l is spread on the surface of an aqueous solution of cadmium chloride of 10^{-4} mole/l. A substrate (measuring $20 \times 30 \times 2$
 30 mm^3) on which transparent conductive layer 22 was formed was degreased and cleaned by applying neutral detergent, trichloroethylene and acetone in the order mentioned. The aforesaid film composed of a mixture of arachic acid and TTF which had been compressed with a surface pressure of 30 dyne/cm was laminated to a 5-ply mass by vertical dipping, thereby providing first organic film 23.

Later, the aforesaid film consisting of a mixture of arachic acid and TTF was completely removed from
 35 the water surface by sucking. Then the arachic acid alone was spread on the water surface. Thereafter, one-ply organic insulation layer 23 composed of arachic acid alone was formed by the vertical dipping process. At the time of lamination, the surface of the arachic acid film was subjected to a pressure of 35 dyne/cm.

After the arachic acid was sucked off the water surface, 100 μ l of the solution was spread over the water surface which was prepared by dissolving 500 mg of arachic acid and 250 mg of 7,7,8,8-tetracyanoquinodimethane (TCNQ) in 100 ml of a solvent prepared by mixing chloroform and acetonitrile in
 40 the volumetric ratio of 1:1. A layer formed of a mixture of arachic acid and TTF was laminated to a 5-ply mass on the laminated mass prepared up to this point by applying a surface pressure of 30 dyne/cm, thereby providing second organic layer 24.

A super lattice structure illustrated in Fig. 3 was fabricated by repeating the above-mentioned step. An
 45 aqueous solution of cadmium used in the spreading step taken in the formation of all the aforesaid Langmuir Blodgett films was held at a temperature of $18 \pm 0.1^\circ C$. A substrate bearing the super lattice structure prepared from said LB films was left for 5 hours in a dry box containing a desiccating agent of silica gel for sufficient drying. Thereafter gold electrode 26 was deposited on said dry laminated mass.

Fig. 4 is a typical band pattern of the surface lattice structure of a display element fabricated in the
 50 aforementioned manner. First organic film 23 containing TTF as donor molecules has a low ionization potential I_{pD} . Second organic film 24 containing TCNQ as acceptor molecules has such electron affinity E_A as is slightly lower than the ionization potential I_{pD} of the first organic film. Insulative organic film 25 has a sufficiently smaller electron affinity and yet a fully higher ionization potential than first and second organic films 23, 24. When, therefore, bias voltage is so impressed as to render first organic film 23 negative, then
 55 the transition of electrons takes place from first organic film 23 to second organic film 24 as indicated by a broken line arrow in Fig. 4. As a result, the light absorbing property of the super lattice structure is changed to ensure a display function.

Description may now be made of a concrete display function. Power source was so connected as to

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render the transparent conductive film 22 of Fig. 3 negative, and gold electrode 26 thereof positive. Light beams of an incandescent lamp were projected from the side of gold electrode 26. When no voltage was impressed, the display element indicated a yellow color. When 15 volts were impressed, the color changed to green. Measurement was made of the response speed at which colors varied with the magnitude of the impressed voltage. It was disclosed that the indication and extinction of a green light followed voltage charges with a delay of 1 ms. 10^5 times of color display operations give rise to no change in the color displaying property of the subject color displaying element.

A color display element was manufactured under the substantially same conditions as in the foregoing embodiment, except that TTF was replaced by tetrathiatetracene (TTT). When 12 volts were impressed, this display element has its yellow color changed to a green color. The response speed at which the color varied with the voltage was 1.2 ms. 10^5 times of color display operations did not cause the display element to make any change in property.

Example 2

Fig. 5 sets forth the case where a series of first and second films 33, 34 were laminated with no insulation film interposed therebetween. This structure indicated the same property as Example 1.

Example 3

Fig. 6 shows the case where laminated bodies of first and second films 43, 44 were stacked one upon another with an insulating film interposed therebetween. This structure also realized the same property as Example 1.

Example 4

This example represents a concrete display element containing a matrix electrode. As seen from Fig. 7A, ITO film stripes 52 were deposited on quartz glass substrate 51 at an interval of about 100 microns and with a width of 500 microns. As indicated in Fig. 5, first donor molecule-containing film 33 and second acceptor molecule-containing film 34 were alternately laminated ten times. In this case, first donor molecule film 33 consisted of a monomolecular LB film prepared by introducing an alkyl group in TTF. Second acceptor molecule film 34 was prepared from a monomolecular LB film prepared by introducing an alkyl group in TCNQ.

Further as shown in Fig. 7B, Al stripe films 53 were deposited at an interval of about 100 microns and with a width of about 500 microns, in such a manner that said Al stripe films 53 extended in the perpendicular direction as well as in the same direction as that in which ITO films 52 were deposited. In this case, Al films 53 with a thickness of 200 Å were formed by vacuum evaporation by means of a metal mask. A region (500 microns × 500 microns) defined by the overlapping of ITO film 52 on Al film 53 constitutes one picture element. In this case, ITO film 52 corresponds to a signal electrode, and Al film 53 corresponds to a scanning electrode.

In this case the following data were obtained:

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5	The threshold voltage V_{th} required for electron transition from the donor molecule to the acceptor molecule	10 V
10	A length of time t_{th} required for the commencement of electron transfer under the condition in which $V_{th} + \Delta V$ ($\Delta V > 0$) was impressed	1 microsecond
15	A length of time t_m required for the ionic condition (D^+A^-) in which electron transfer took place to regain the original neutral state (D_0A_0) with the 0 voltage impressed	40 milliseconds
20		

In the display element embodying the present invention, the number of picture elements was set at 10,000, and the picture area was defined to have a size of 6 cm x 6 cm.

Description may now be made of the operation of the above-described display element.

Fig. 8A is a chart illustrating the principle on which the start and stop of the display operation of one picture element ($X_i; Y_j$).

Now let it be assumed that pulse voltage V_{xi} (= 9 V) is impressed on scanning electrode X_i for a length of time t_x (= 400 microseconds). During said period t_x , signal voltage V_{yj} is successively impressed on signal electrode Y_j for a length of time t_y (= 4 microseconds). In this case, a picture element desired to display a color has its signal electrode impressed with pulse voltage V_{ON} (= -2.0 V). A picture element required to cease a color display has its signal electrode impressed with voltage V_{OFF} (= 0.0 V). Namely, when the picture element ($X_i; Y_j$) is rendered on, the following steps are taken:

$$V_{xi} = V_x = 9.0 \text{ V}$$

$$V_{yj} = V_{ON} = -2.0 \text{ V}$$

$$\text{Therefore, } V_{xi} - V_{yd} = 11.0 \text{ V} > V_{th} = 10.0 \text{ V}$$

When the picture element is rendered off, the following step is taken:

$$V_{xi} = V_x = 9.0 \text{ V}$$

$$V_{yj} = V_{OFF} = 0.0 \text{ V}$$

$$\text{Therefore } V_{xi} - V_{yd} = 9.0 \text{ V} < V_{th} = 10.0 \text{ V}$$

Once rendered on, a picture element sustains display during a period of time t_m (= 40 milliseconds).

Fig. 8B represents the driving signal timing chart of a matrix display. The scanning electrodes $X_1, X_2 \dots X_n$ are successively impressed with a pulse voltage of 9 V having a width of 400 microseconds. Since 100 scanning electrodes are provided, pulse voltage is impressed with a period of 40 milliseconds. Signal electrodes $Y_1, Y_2 \dots Y_m$ are impressed with signal pulse voltage having a width of 4 microseconds with a period of 400 microseconds. When the signal pulse voltage stands at -2 V, the corresponding element is rendered conducting for display. When the signal pulse voltage indicates 0 V, the corresponding picture element is rendered off. Fig. 8 indicates the on and off states of the respective picture elements.

Example 5

Fig. 9 illustrates the construction of a multicolor display element embodying the present invention. Reference numeral 61 represents a glass substrate. Deposited on this substrate is a transparent conductive film 62 prepared from, for example, ITO. Further, insulative molecule film 64a is deposited on said conductive film 62 with inorganic insulative film 63 interposed therebetween. Formed on said laminated assembly is a surfar lattice structure constructed by alternately laminating acceptor molecule-containing film 66 and donor molecule-containing film 65 which comprises two different donor molecules D_1, D_2 .

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Inorganic insulative film 63 was formed by depositing SiO_2 film by the CVD process with a thickness of 2000 Å. Insulative molecule-containing films 64a, 64b, acceptor molecule-containing film 66 and donor molecule-containing film 65 were prepared by the Langmuir blodgett process. To described concretely, acceptor molecule-containing film 66 was fabricated by providing an alkyl chain derivative of tetracyanoquinodimethane (TCNQ) as an acceptor molecule, mixing said derivative with stearic acid in the molecular ratio of 1:1 and laminated two resultant monomolecular LB films. Donor molecule-containing film 65 was formed by providing an alkyl chain derivative of tetrathiafulvalene (TTF) and an alkyl chain derivative of paraphenylene diamine (PD) as donor molecules, mixing said two alkyl chain derivatives with stearic acid in the molar ratio of 1:1:2 (2 represents the content of stearic acid), and laminated the two resultant monomolecular LB films. Insulative molecule-containing films 64a, 64b were formed by laminating five stearic LB films. Top electrode 67 was fabricated by the evaporation of aluminium with a thickness of 100 Å.

When said top electrode 67 is impressed with such voltage as to be rendered positive, then a white transmitted spectrum changes as mentioned below in accordance with the level of said voltage. Fig. 10 indicates said spectrum changes. Notation ① indicates a pattern of a transmitted spectrum appearing when no voltage is impressed. When the impressed voltage rises higher than $V_{tr}(1) = 1.5$ V, the transmitted spectrum shows a pattern represented by notation ②. Further when the impressed voltage exceeds $V_{tr}(2) = 2.5$ V, the transmitted spectrum presents a pattern marked by notation ③. Accordingly, a color to be displayed can be changed in 3 steps in accordance with the level of the impressed voltage as illustrated below:

In case of $V < V_{tr}(1)$	yellow
In case of $V_{tr}(1) < V < V_{tr}(2)$	blue
In case of $V_{tr}(2) < V$	red

In the above-mentioned display element representing the present invention, the organic films can be laminated in any optional number of plies. Further, the thickness of the insulative film 63 may have any optional thickness. Proper selection of the number of the laminated films and the thickness of the respective films ensures the identical property of the basic display element, though minor changes appear in threshold voltage $V_{tr}(1)$, $V_{tr}(2)$.

Example 6

This example is represented by another type of multicolor display element illustrated in Fig. 11.

The multicolor display element of Fig. 11 has a different arrangement from that of Fig. 9 in the under-mentioned respect. Namely, the organic film has a hetero structure represented by film 68 consisting of two donor molecules D1, D2 and one acceptor molecule A. Said film mixture is represented by an LB film which is prepared by mixing an alkyl chain derivative of TTF acting as a donor molecule, alkyl chain derivative of PD, alkyl chain derivative of TCNQ acting as an acceptor molecule and stearic acid acting as an insulating molecule in the molar ratio 1:1:2:2. According to the present invention, the laminated film body 8 is defined to consist of, for example, 14 plies.

Example 6 ensures the manufacture of a color display element capable of indicating a large number of colors like the preceding Example 5.

Example 7

This example refers to still another type of multicolor display element (Fig. 12). Example 7 represents two hetero super lattice structures. This assembled body is composed of one super lattice structures prepared by laminating a large number of hetero structure respectively consisting of donor molecule films 65a containing donor molecule D₁ and acceptor molecule films 66 and another super lattice structure constructed by laminating a large number of hetero layer structures composed of donor molecule film 65b containing donor molecule D₂ and acceptor molecule film 66. In Example 7, one donor molecule film 65a consists of an LB film prepared by film 65a consists of an LB film prepared by mixing TTF with stearic acid is the molar ratio of 1:1. Another donor molecule film 65b consists of an LB film obtained by mixing PD and stearic acid in the molar ratio of 1:1. Example 7 of Fig. 12 indicates the same satisfactory properties as those of the preceding examples.

In the foregoing Examples 5 to 7, two kinds of donor molecules and one kind of acceptor molecule were

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assembled together. However more kinds of donor and acceptor molecules may be assembled, thereby ensuring the display of more than three colors. Further, various kinds of organic films described in Examples 5-7 may be properly assembled.

In Examples 5-7, inorganic insulative film 63 may be omitted. Similarly, insulative molecule-containing films 64a, 64b and 64c may be omitted, in case it is possible to build a barrier sufficiently effective to obstruct the electric charge transfer between an organic film and electrode 67 or transparent conductive film 62 deposited on the substrate by selection of proper materials. However, insulative molecule-containing films 64a, 64b and 64c are indispensable in order to ensure more effective charge transfer by reliably preventing carriers from being taken into an organic film from an electrode or substrate. Further as viewed from the stability of the properties of elements, it is required to provide the insulative molecule-containing films 64a, 64b and 64c particularly the insulative molecule-containing film 64b, 64c positioned underneath electrode 67. The reason is as follows. Generally, a method electrode indicates by itself a strong acceptor or donor characteristic, and also is possessed of high diffusivity. If, therefore, the metal electrode is brought into direct contact with a donor molecule-containing film or acceptor molecule-containing film, then a complex compound tends to be readily produced, probably resulting in the deterioration of the property of display element.

Throughout the foregoing examples 5-7, TTF and PD were cited as donor molecules, TCNQ as an acceptor molecule and stearic acid as an insulative molecule. Since, however, a light-absorbing property varies with the kind of an organic molecule, the subject display element can indicate diversified colors by properly selecting the kind of molecules to be applied.

Example 8

This example shows a concrete display element having a matrix electrode. The element had the hetero-structure shown in Fig. 12. Its matrix electrode was identical with that illustrated in Figs. 7A and 7B. Its film D₁ was a monomolecular LB film of an alkyl chain derivative of TTF. Its film D₂ was a monomolecular LB film of PD. Its film A was a monomolecular film of an alkyl chain derivative of TCNQ. The element was comprised of ten units each consisting of film D₁ and film A, and ten units each consisting of film D₂ and film A.

The display element emitted yellow light when a voltage of less than 16 v was applied to it. It turned blue when a voltage ranging from 16 v to 20 v was applied to it. It glowed red when a voltage of more than 20 v was applied to it. About 1-microsecond had passed until the element displayed these colors after the application of the voltage. Molecules D₁⁺A⁻ and D₂⁺A⁻ required about 40 milliseconds to become electrically neutral after the application of voltage had been stopped.

The operation of the one-picture elements forming the display element of Example 8 will be explained with reference to Fig. 13A. The one-picture element consisting of a scanning electrode X_i and a signal electrode Y_j glows red when -8 V is applied to electrode Y_j while +14 V is being applied to electrode X_j. This one-picture element turns blue when -4 V is applied to signal electrode Y_j in the same condition. The one-picture element turns yellow when 0 V is applied to signal electrode Y_j in the same condition. Each of these display condition stays unchanged for approximately 40 milliseconds after the application of the voltage to electrode Y_j.

Fig. 13B is a timing chart showing when driving signals are supplied to the matrix display element of Example 8. The timing of supplying the driving signals is the same as in the mono-color display of Example 4, but Example 8 is different from Example 4 in that signal pulse voltages of 0 V, -4 V and -8 V are applied to the one-picture elements, thereby causing the one-picture elements to emit yellow light, blue light and red light, respectively.

Claims

1. An organic thin film display element comprising:
 - an organic thin film containing at least two parallel organic monomolecular layers, one (13) of which contains organic molecules of at least the donor kind, the other (14) of which contains organic molecules of at least the acceptor kind;
 - a pair of electrodes (11, 15) parallel to said thin film, said thin film being positioned between said electrodes; at least a first additional monomolecular layer (12a) containing insulating organic molecules and being positioned between one of said electrodes (11) and said thin film; at least a second additional monomolecular layer (12b) containing insulating organic molecules and being positioned between the other (15) of said electrodes and said thin film;

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said electrodes (11, 15) being arranged to cause a charge transfer between the donor molecules and the acceptor molecules, said charge transfer varying the optical transmission of the organic thin film, whereby the organic thin film displays an image.

- 5 2. A display element according to claim 1, characterized in that said film comprises a plurality of organic monomolecular layers (23) containing organic molecules of at least the donor kind which layers alternate with organic monomolecular layers (24) which contain organic molecules of at least the acceptor kind.
- 10 3. A display element according to claim 1 or claim 2, characterized in that the or each layer containing organic molecules of at least the donor kind contains donor molecules of different kinds.
4. A display element according to any of claims 1 to 3, characterized in that the or each layer which contains organic molecules of at least the acceptor kind contains acceptor molecules of different kinds.
- 15 5. A display element according to any of claims 1 to 4, characterized in that a further monomolecular layer (25) containing insulating organic molecules is interposed between the or a layer (23) containing organic molecules of at least the donor kind and the or an adjacent layer containing organic molecules of at least the acceptor kind.
- 20 6. The display element according to any of claims 1 to 5, characterized in that said organic thin film is of super lattice structure.
- 25 7. A display element according to claim 1, characterized in that the electrodes are stripe-like scanning and signal electrodes respectively and are at right angles to each other and in that means are connected to the scanning electrode and signal electrode for selectively applying a pulse voltage to said organic thin film.

Revendications

- 30 1. Elément d'affichage à film mince organique comprenant :
un film mince organique contenant au moins deux couches monomoléculaires organiques parallèles, dont l'une (13) contient des molécules organiques du type donneur au moins et dont l'autre (14) contient des molécules organiques du type accepteur au moins ;
35 une paire d'électrodes (11, 15) parallèles audit film mince, ledit film mince étant situé entre lesdites électrodes ; au moins une première couche monomoléculaire supplémentaire (12a) contenant des molécules organiques isolantes et située entre l'une (11) desdites électrodes et ledit film mince ; au moins une seconde couche monomoléculaire supplémentaire (12b) contenant des molécules organiques isolantes et située entre l'autre (15) desdites électrodes et ledit film mince ;
40 lesdites électrodes (11, 15) étant agencées de manière à provoquer un transfert de charges entre les molécules donneuses et les molécules acceptrices, ledit transfert de charges modifiant la transmission optique du film mince organique de sorte que le film mince organique affiche une image.
2. Elément d'affichage selon la revendication 1, caractérisé en ce que ledit film comprend une multiplicité
45 de couches monomoléculaires organiques (23) contenant des molécules organiques du type donneur au moins, ces couches alternant avec des couches monomoléculaires organiques (24) qui contiennent des molécules organiques du type accepteur au moins.
3. Elément d'affichage selon la revendication 1 ou la revendication 2, caractérisé en ce que la ou chaque
50 couche contenant des molécules organiques du type donneur au moins contient des molécules donneuses de différents types.
4. Elément d'affichage selon l'une quelconque des revendications 1 à 3, caractérisé en ce que la ou
55 chaque couche qui contient des molécules organiques du type accepteur au moins contient des molécules acceptrices de différents types.
5. Elément d'affichage selon l'une quelconque des revendications 1 à 4, caractérisé en ce qu'une autre
couche monomoléculaire (25) contenant des molécules organiques isolantes est interposée entre la ou

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une couche (23) contenant des molécules organiques du type donneur au moins et la ou une couche adjacente contenant des molécules organiques du type accepteur au moins.

6. Élément d'affichage selon l'une quelconque des revendications 1 à 5, caractérisé en ce que ledit film mince organique a une structure de type surstructure.
7. Élément d'affichage selon la revendication 1, caractérisé en ce que les électrodes sont des électrodes de balayage et de signal respectivement en forme de bandes qui sont perpendiculaires entre elles et en ce que des dispositifs sont reliés à l'électrode de balayage et à l'électrode de signal pour appliquer sélectivement une tension en impulsions audit film mince organique.

Patentansprüche

1. Organisches Dünnschicht-Bildwiedergabeelement, umfassend:
 - eine organische Dünnschicht mit mindestens zwei parallelen organischen monomolekularen Schichten, von denen eine (13) organische Moleküle zumindest der Donorart und die andere (14) organische Moleküle zumindest der Akzeptorart enthält;
 - ein parallel zu der Dünnschicht ausgerichtetes Elektrodenpaar (11, 15), wobei sich die Dünnschicht zwischen den Elektroden befindet; mindestens eine erste zusätzliche monomolekulare Schicht (12a) mit isolierenden organischen Molekülen, die sich zwischen einer der Elektroden (11) und der Dünnschicht befindet; mindestens eine zweite zusätzliche monomolekulare Schicht (12b) mit isolierenden organischen Molekülen, die sich zwischen der anderen (15) der Elektroden und der Dünnschicht befindet; wobei die Elektroden (11, 15) derart angeordnet sind, daß sie eine Ladungsübertragung zwischen den Donormolekülen und den Akzeptormolekülen herbeiführen, wobei die Ladungsübertragung die optische Durchlässigkeit der organischen Dünnschicht unter Darstellung eines Bildes durch dieselben variiert.
2. Bildwiedergabeelement nach Anspruch 1, dadurch gekennzeichnet, daß der Film eine Mehrzahl von organischen monomolekularen Schichten (23) mit organischen Molekülen zumindest der Donorart umfaßt, wobei sich diese Schichten mit organischen monomolekularen Schichten (24) mit organischen Molekülen zumindest der Akzeptorart abwechseln.
3. Bildwiedergabeelement nach Anspruch 1 oder 2, dadurch gekennzeichnet, daß die oder jede Schicht mit organischen Molekülen zumindest der Donorart Donormoleküle verschiedener Arten enthält.
4. Bildwiedergabeelement nach einem der Ansprüche 1 bis 3, dadurch gekennzeichnet, daß die oder jede Schicht mit organischen Molekülen mindestens der Akzeptorart Akzeptormoleküle unterschiedlicher Arten enthält.
5. Bildwiedergabeelement nach einem der Ansprüche 1 bis 4, dadurch gekennzeichnet, daß zwischen der oder einer Schicht (23) mit organischen Molekülen mindestens der Donorart und der oder einer benachbarten Schicht mit organischen Molekülen zumindest der Akzeptorart eine weitere monomolekulare Schicht (25) mit isolierenden organischen Molekülen eingefügt ist.
6. Bildwiedergabeelement nach einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß der organische Dünnschicht Supergitterstruktur aufweist.
7. Bildwiedergabeelement nach Anspruch 1, dadurch gekennzeichnet, daß die Elektroden aus streifenförmigen Abtast- bzw. Signalelektroden bestehen und zueinander in rechten Winkeln angeordnet sind und daß an die Abtastelektrode und Signalelektrode zum selektiven Anlegen eines Stromimpulses an den organischen Dünnschicht eine Einrichtung angeschlossen ist.

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FIG. 1

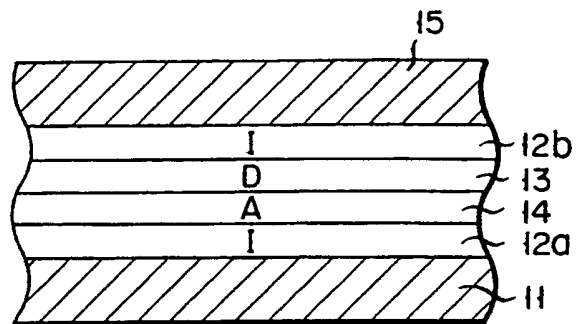


FIG. 2

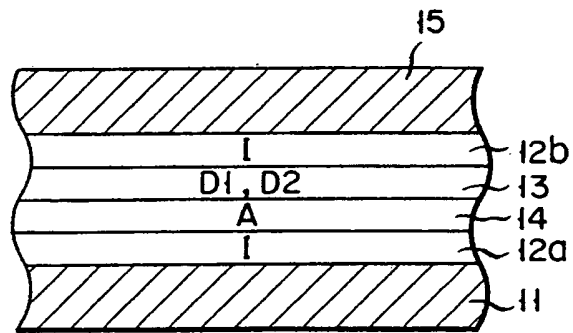
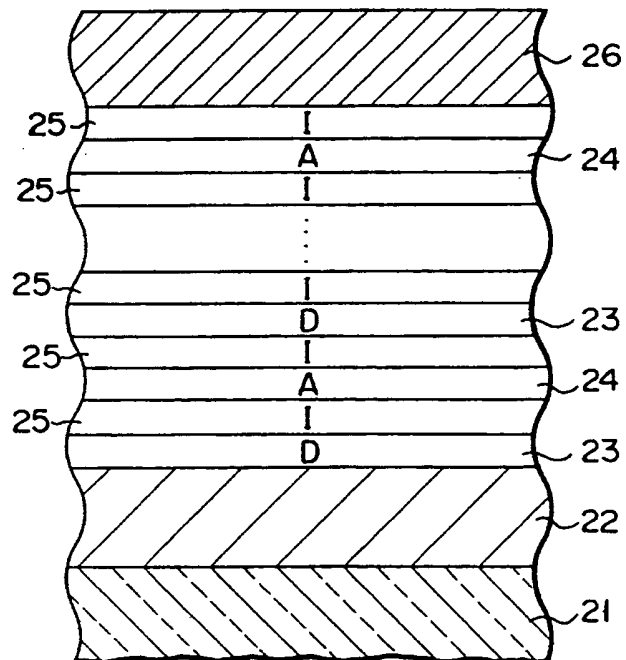


FIG. 3



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FIG. 4

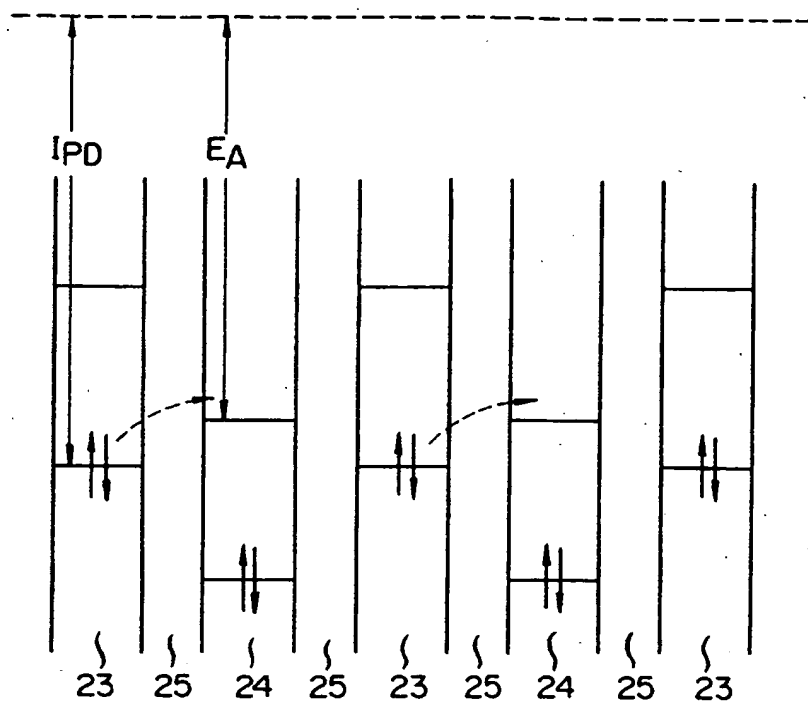


FIG. 5

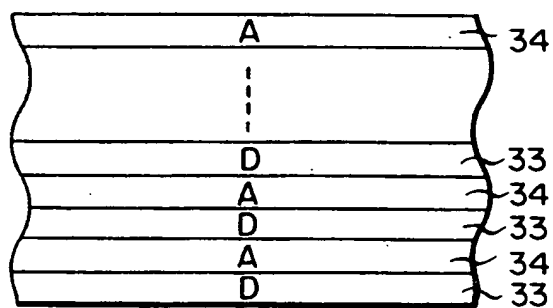
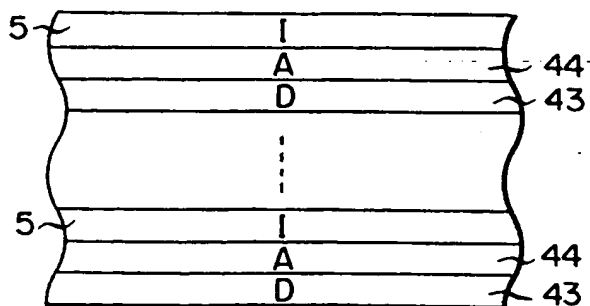


FIG. 6



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FIG. 7A

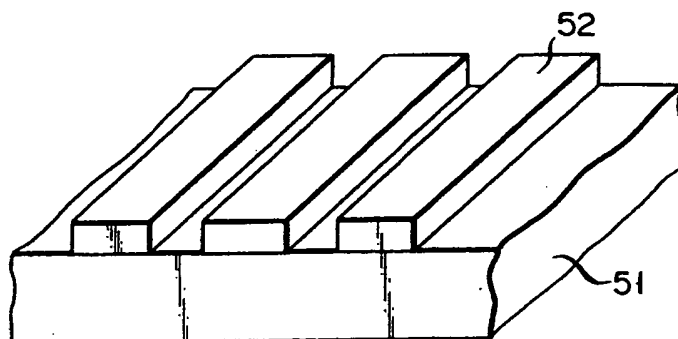
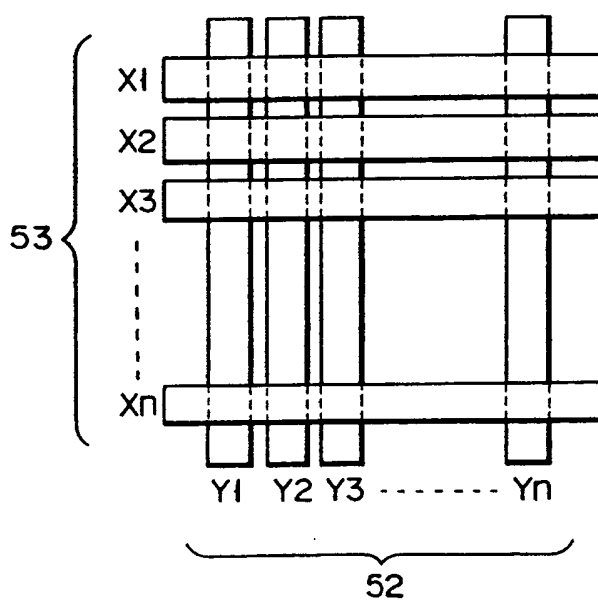


FIG. 7B



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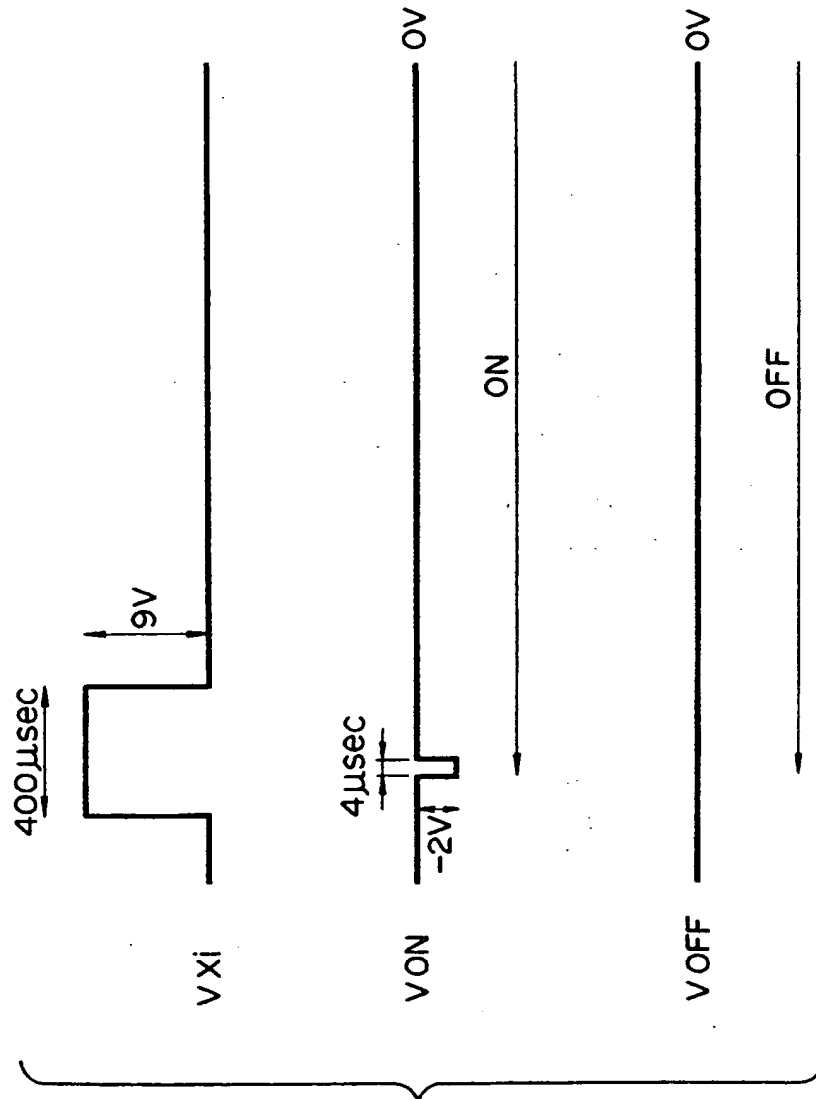
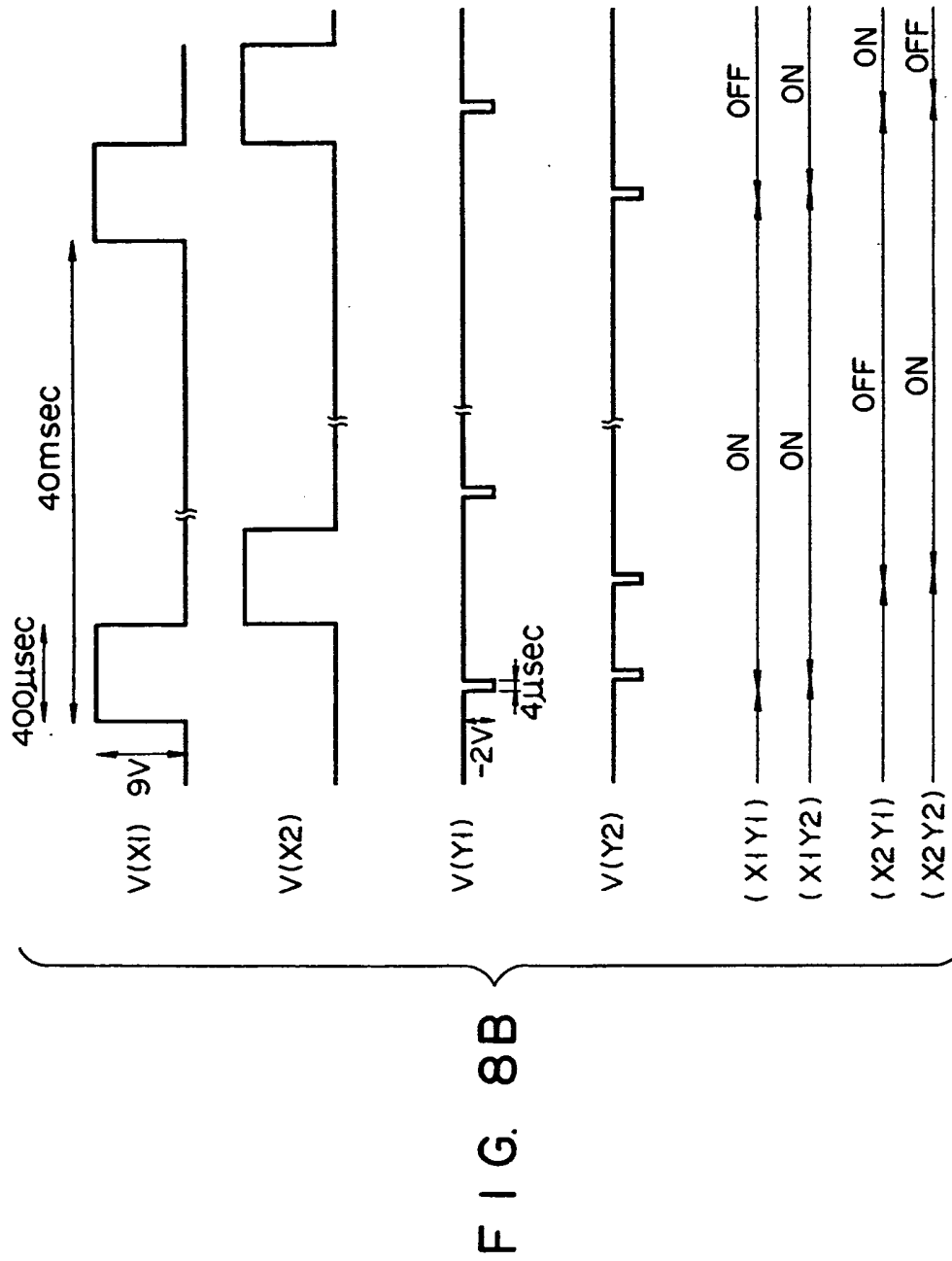


FIG. 8A

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FIG. 9

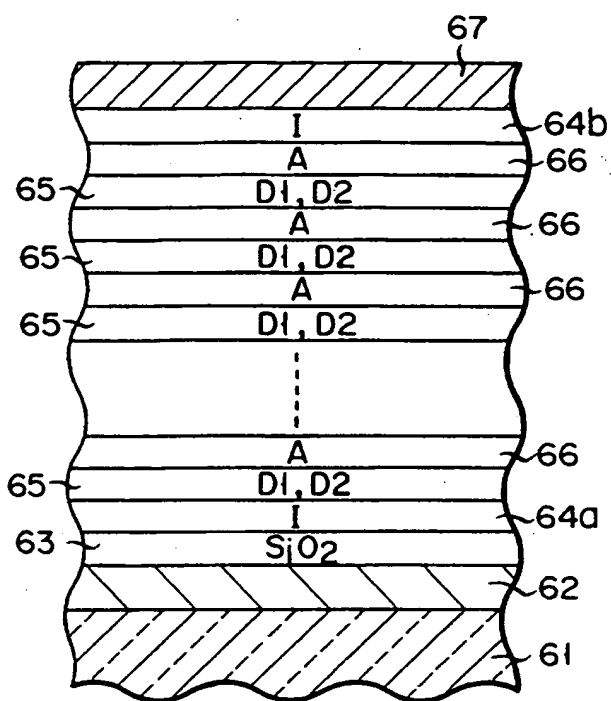
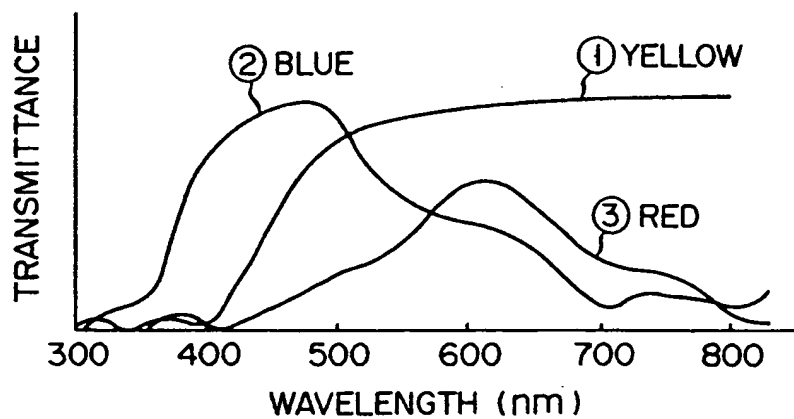


FIG. 10



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FIG. 11

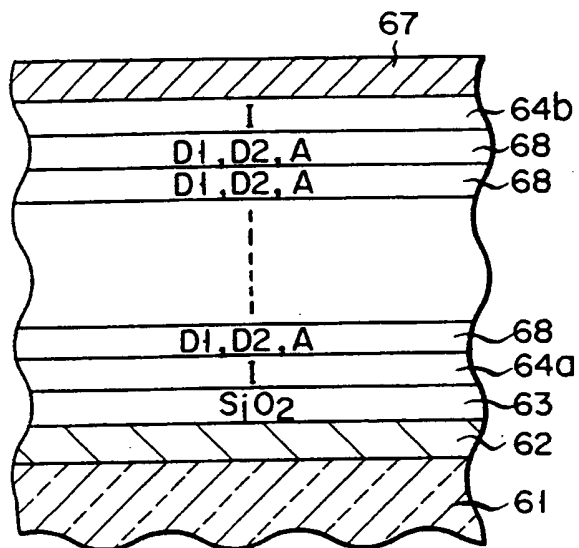
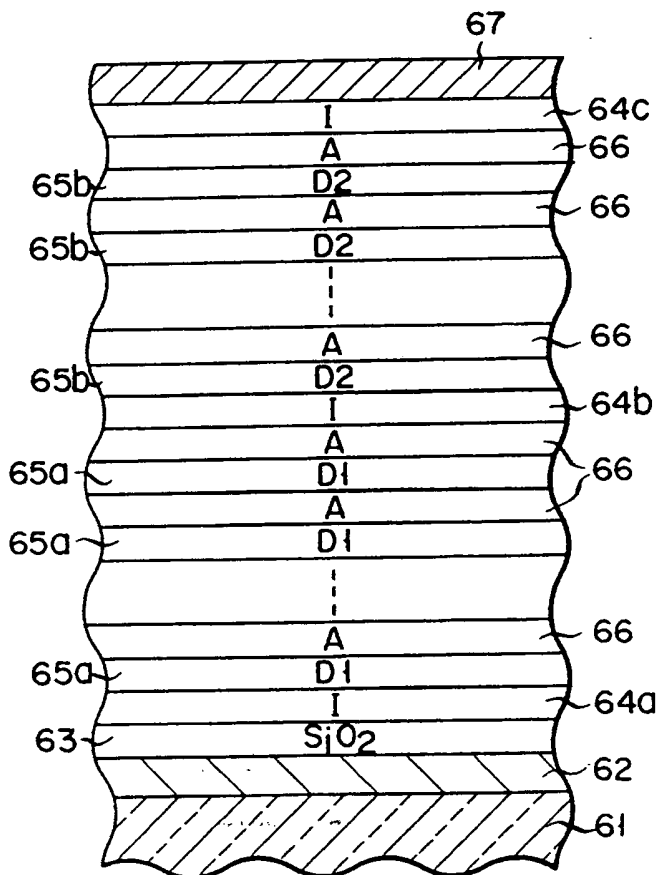
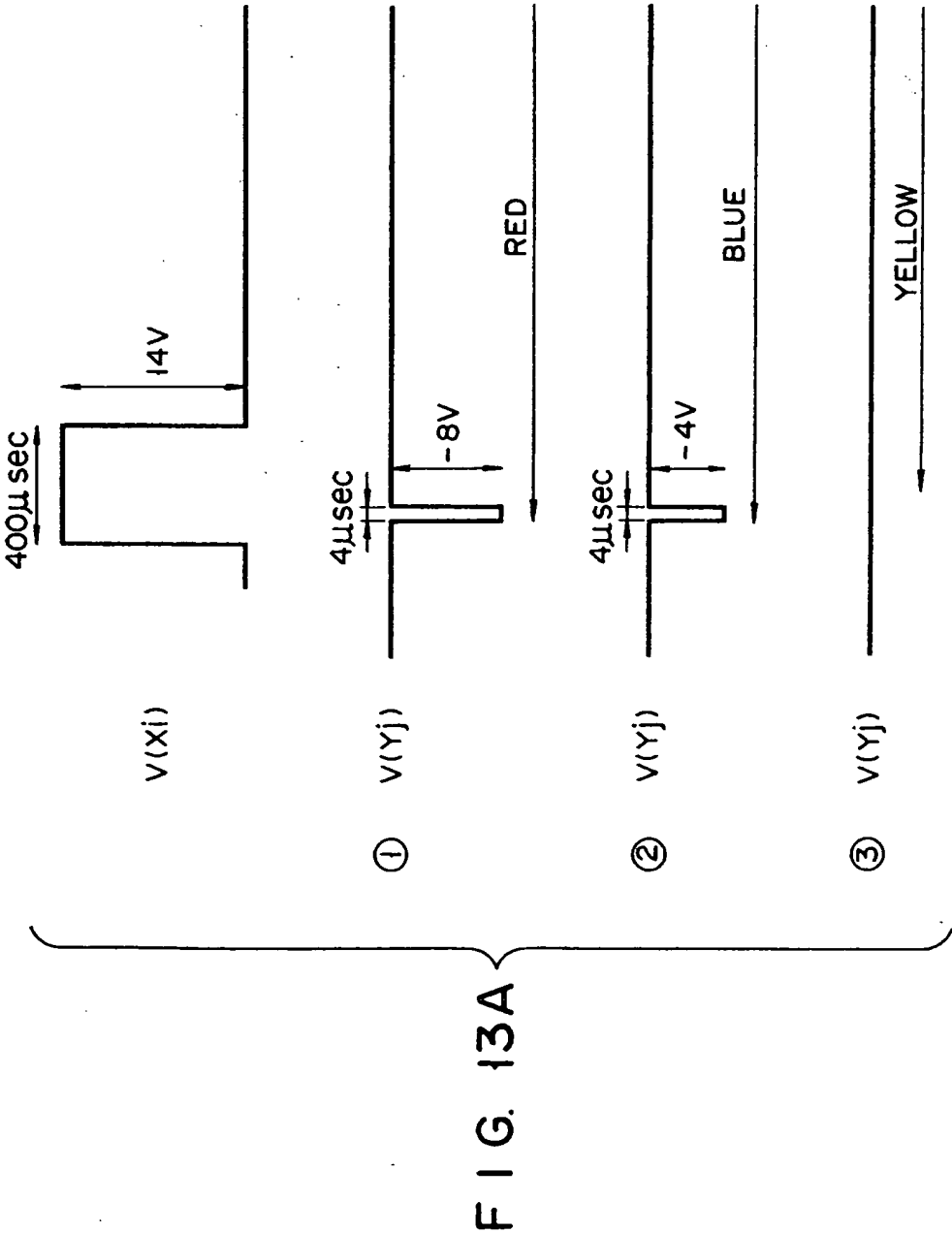


FIG. 12

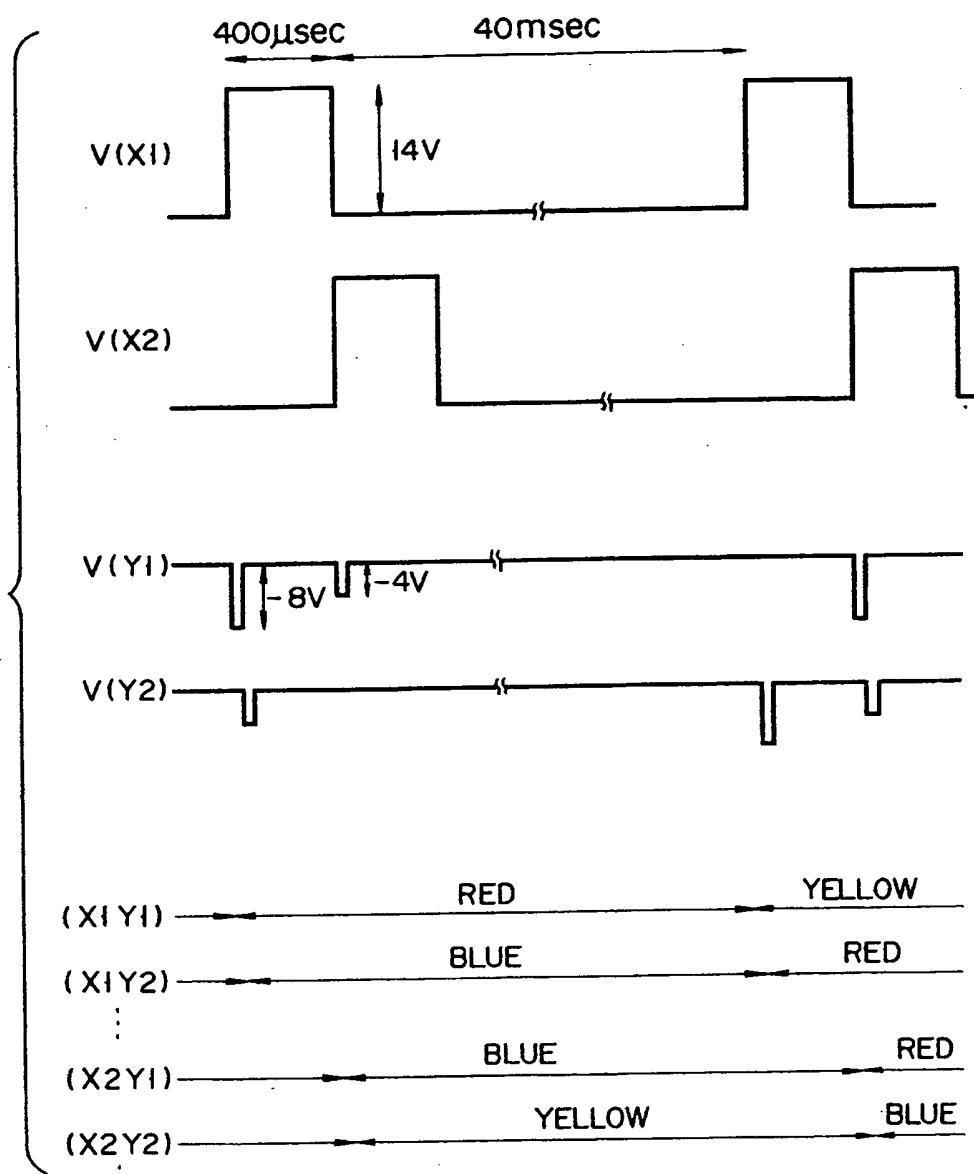


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FIG. 13B



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